



WW LCI v2: A second-generation life cycle inventory model for chemicals discharged to wastewater systems

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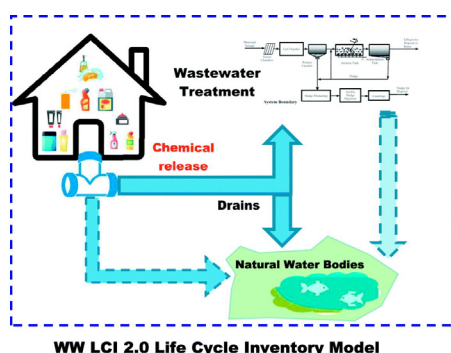
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HIGHLIGHTS

- Second-generation wastewater treatment inventory model, WW LCI 2.0 is developed.
- A novel approach for quantifying the complete inventories for waste treatment.
- WW LCI 2.0 takes into account different wastewater treatment levels.
- Higher treatment levels lead to lower CC and FET burden compared to direct discharge.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 27 July 2017

Received in revised form 6 October 2017

Accepted 6 October 2017

Available online 18 October 2017

Editor: D. Barcelo

Keywords:

Environmental fate model

Life cycle inventory

Life cycle assessment

Sewage

Wastewater

WW LCI

ABSTRACT

We present a second-generation wastewater treatment inventory model, WW LCI 2.0, which on many fronts represents considerable advances compared to its previous version WW LCI 1.0. WW LCI 2.0 is a novel and complete wastewater inventory model integrating WW LCI 1.0, i.e. a complete life cycle inventory, including infrastructure requirement, energy consumption and auxiliary materials applied for the treatment of wastewater and disposal of sludge and SewageLCI, i.e. fate modelling of chemicals released to the sewer. The model is expanded to account for different wastewater treatment levels, i.e. primary, secondary and tertiary treatment, independent treatment by septic tanks and also direct discharge to natural waters. Sludge disposal by means of composting is added as a new option. The model also includes a database containing statistics on wastewater treatment levels and sludge disposal patterns in 56 countries. The application of the new model is demonstrated using five chemicals assumed discharged to wastewater systems in four different countries. WW LCI 2.0 model results shows that chemicals such as diethylenetriamine penta (methylene phosphonic acid) (DTPMP) and Diclofenac, exhibit lower climate change (CC) and freshwater ecotoxicity (FET) burdens upon wastewater treatment compared to direct discharge in all country scenarios. Results for Ibuprofen and Acetaminophen (more readily degradable) show that the CC burden depends on the country-specific levels of wastewater treatment. Higher treatment levels lead to lower CC and FET burden compared to direct discharge. WW LCI 2.0 makes it possible to generate complete detailed life cycle inventories and fate analyses for chemicals released to wastewater systems. Our test of the WW LCI 2.0 model with five chemicals illustrates how the model can provide substantially different outcomes, compared to conventional wastewater inventory models, making the inventory dependent upon the atomic composition of the molecules undergoing treatment as well as the country specific wastewater treatment levels.

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1. Introduction

Wastewater is one of the main contributors to global environmental pollution. Whenever wastewater is emitted without adequate treatment, the untreated/partially treated wastewater pose various environmental impact potentials, such as marine and freshwater eutrophication and ecotoxicity. Although wastewater treatment can be the solution, treating wastewater is a resource-demanding activity, involving use of, among others, energy and chemicals. There are numerous approaches and technologies by which wastewater treatment can be achieved. However, to make wastewater treatment sustainable there is a need for more holistic/systemic assessments of wastewater treatment technologies. Life Cycle Assessment (LCA) has by now become a widely adopted approach for assessing environmental sustainability in the wastewater treatment sector (Corominas et al., 2013). A common foundation for including wastewater treatment in LCA studies is the Ecoinvent database, where inventories for wastewater treatment sourced in Switzerland are included. The Ecoinvent database uses the wastewater life cycle inventory (LCI) model for Switzerland developed by (Doka, 2007). These Ecoinvent inventories add uncertainty to the wastewater LCA given that the data on energy consumption, chemical consumption and infrastructures (wastewater and sewer) are specific for the location for which an inventory is generated, i.e. Switzerland in the Ecoinvent case, and yet they are used as representative inventories for many other countries in the world.

In addition, there is a growing content of trace pollutants in wastewater because of the ever increasing use of chemicals such as Pharmaceuticals and Personal Care Products (PPCP) (Muñoz et al., 2008; Jiang et al., 2013; Luo et al., 2014). A considerable fraction of such chemicals (pharmaceuticals, disinfectant etc.) are designed to have high biological activities are hence likely to cause effects in the environment upon emission. Complex organic molecules all behave differently in wastewater treatment systems and pose different resource demands than more ordinary wastewater constituents such as inorganic chemicals (Muñoz et al., 2016). In order to correctly account for the environmental damages posed by these micro-pollutants upon emission to the environment even after wastewater treatment, there is a need for establishing dedicated life cycle inventories accounting for the resources needed to remove or more correctly lower the concentration of chemicals in wastewater upon treatment, as well as the particular fraction of the chemicals passing through the wastewater system and ending up in the environment via various elementary flows. The Ecoinvent model proves unsuitable to account for the diversity of chemical behavior in wastewater treatment systems since it assumes the same biodegradation coefficients (app. 53%) for all types of organic chemicals (Doka, 2007, p. 18). Such simplification is inappropriate in the case of micro-pollutants such as e.g. PPCPs and for many other chemicals found in wastewater (Clark et al., 1995; Joss et al., 2006; Kujawa-Roeleveld and Schuman, 2008). This highlights the need to develop dedicated inventories for assessing the environmental burdens associated with micro-pollutants emitted to wastewater systems.

Recently, there have been efforts in the direction of developing inventory models for micro-pollutants in wastewater. The model SewageLCI (Birkved and Dijkman, 2012) was developed focusing on fate and transport of micro-pollutants in wastewater systems. The model provides the fraction of a chemical emitted to wastewater systems and subsequently undergoing fate processes in country/site specific steps of wastewater treatment (i.e. primary, secondary and tertiary treatment). A complete mass balance of the chemical being emitted to the wastewater system is provided as well as all the remaining fractions, such as the fraction emitted to air, surface water recipients and the fraction transferred to sludge. However, the SewageLCI model does not provide the complete inventory of the wastewater systems i.e. the inventory of the infrastructure required for wastewater treatment, energy consumption and other materials used during the treatment.

Muñoz et al. (2016) developed WW LCI 1.0, a model that provides the complete inventory of wastewater treatment systems. This model takes into account infrastructure requirements, energy consumption and auxiliary chemicals used for the treatment as well as sludge treatment including disposal hereof. The model also performs a mass balance of the chemicals emitted to the wastewater system. A limitation of this model is that it only considers wastewater treatment at secondary treatment level. Hence the model does not reflect the way in which the actual wastewater treatment systems are constructed and operated in most real-life situations. These past efforts suggest that although there have been attempts to provide inventory models for wastewater treatment for specific chemicals, none of these models are comprehensive, complete in nature and hence unable to properly provide real-life inventories.

In this paper, we present and demonstrate the WW LCI 2.0 model, which integrates the approach of SewageLCI (i.e. fate modelling of chemical released to the sewer), with the complete life cycle inventory (i.e. the inventory of infrastructure requirement, energy consumption and chemical used for the treatment of wastewater and disposal of sludge) approach of WW LCI 1.0. The developed integrated model addresses major limitations of past models, thus representing a substantial step forward. In addition, the model comes with a comprehensive country database on the wastewater treatment levels and practice of sludge handling applied in 56 countries.

2. Methodology

WW LCI 2.0 builds on the calculation modules of WW LCI 1.0. The reference flow for all the calculations remains unchanged i.e. “1 kg chemical discharged down the drain”. Table 1 shows the differences between the WWLCI 1.0 and WW LCI 2.0 model. As it can be seen from this table there are various new developments made in WW LCI 2.0 to improve the existing WW LCI 1.0 model. The new model presented here nevertheless differs from WW LCI 1.0 in the chosen approach for modelling of Wastewater Treatment Plants (WWTPs). Instead of considering only a standard WWTP with primary and secondary (biological) treatment, WW LCI 2.0 goes beyond this and further accounts for different levels of treatment. This approach of generating emissions inventory at treatment level was followed in Sewage LCI as reported in Birkved and Dijkman (2012). WW LCI 2.0 basically incorporates the features of WW LCI 1.0 and SewageLCI. Fig. 1 shows the various components considered in the model and different percentages of wastewater along with the fractions of a chemical that are available as an output from the model. Fig. 1 also shows that WW LCI 2.0 takes into account the direct discharge of wastewater to surface water and independent treatment in septic tanks, followed by disposal of septage.

In WW LCI 2.0, the input data requirement remains the same as in WW LCI 1.0, with only one additional chemical property required, namely the sludge-water partition coefficient (K_d). Further, all calculations relating to biological treatment (organic matter removal, N removal), P removal, sludge digestion, energy use calculations and heat generation from sludge-derived biogas are the same as in the earlier version of the model.

Degradation of chemicals in the environment after the release to natural bodies was taken into account in WW LCI 1.0, according to the pathways described in Muñoz et al. (2013). This approach of generating inventories for chemicals (i.e. with inclusion of environmental degradation) is retained in WW LCI 2.0. However, as degradation takes place in the ecosphere, and should be captured in impact assessment calculations rather than in the inventory analysis (Muñoz et al., 2013), in WW LCI 2.0 an additional option of creating inventory without accounting for environmental degradation is provided. Users can choose this inventory generation option according to the system boundary preferences.

Table 1
New developments in WW LCI 2.0 and major differences with WW LCI 1.0.

			WW LCI v1	WW LCI v2
Wastewater treatment levels	WWTP with primary treatment only		✗	✓
	WWTP with primary & secondary treatment	With N removal	✓	✓
		Without N removal	✓	✓
	As above, plus tertiary treatment (sand filter)		✗	✓
	Chemical phosphorus removal		✓	✓
Sludge disposal	Septic tanks		✗	✓
	No treatment (direct discharge)		✓	✓
	Composting		✗	✓
	Landfarming		✓	✓
	Landfilling		✓	✓
Database	Incineration		✓	✓
	Wastewater treatment statistics		✗	✓
	Sludge disposal statistics		✗	✓
LCI results	Exportable to SimaPro		✓	✓
	Possibility of excluding environmental degradation emissions		✗	✓

The following sub-sections describe in detail the new features of WW LCI 2.0, namely the underlying data sources, calculations and assumptions.

2.1. WWTP with primary treatment only

WWTPs applying only primary treatment have been modeled as a conventional sedimentation tank. The main purpose of primary treatment is to remove suspended solids. Primary treatment typically achieves 30% Biochemical Oxygen Demand (BOD) removal efficiency and 30% removal of Suspended Solids (SS) in primary sludge (Metcalf et al., 2007). Using the same approach, the degradation and sorption to sludge rates for a given chemical are calculated in accordance with

Eqs. (1) and (2):

$$F_{deg.pt} = r_{deg.pt} \times F_{deg.as} \quad (1)$$

$$F_{sludge.pt} = r_{sludge.pt} \times F_{sludge.as} \quad (2)$$

where, $F_{deg.pt}$ is the fraction of chemical biodegradation in WWTPs with primary treatment only, $F_{deg.as}$ is the fraction of biodegradation in full-fledged WWTPs (primary treatment followed by activated sludge process), $r_{deg.pt}$ is the ration (expressed in percentage) of degradation efficiency in WWTPs with only primary treatment to degradation efficiency in full scale WWTPs (typically considered as 30%).

$F_{sludge.pt}$ is the fraction of solids removal in WWTPs with primary treatment only, $F_{sludge.as}$ is the fraction of solids removed in full-

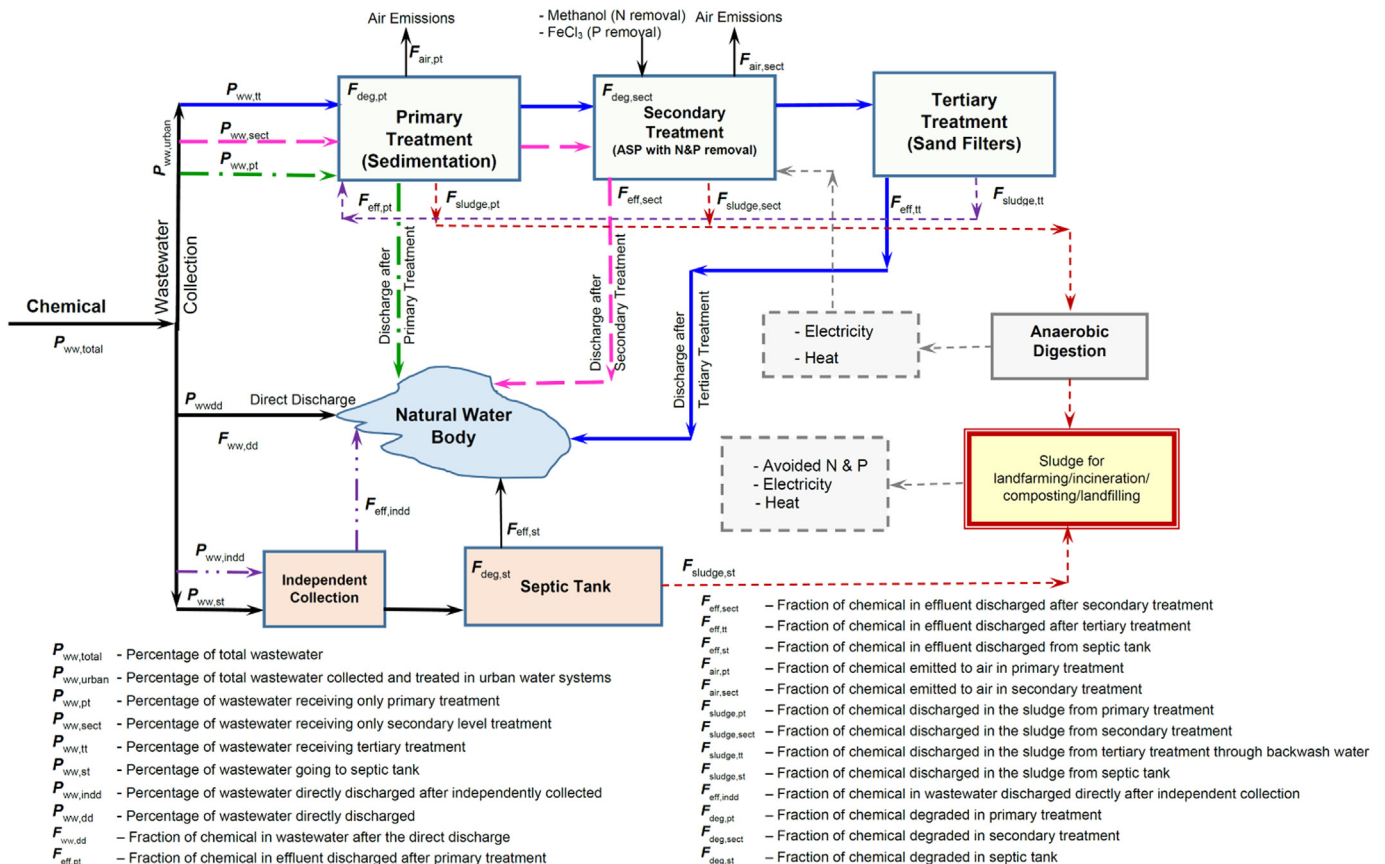


Fig. 1. Flow scheme for WW LCI 2.0.

fledged WWTPs, $r_{sludge,pt}$ is the ration (expressed in percentage) of SS removal efficiency in WWTPs with only primary treatment to SS removal efficiency in full scale WWTP (typically considered as 30%).

In this way, removal rates of a given chemical in a WWTP with only primary treatment are thus linked to the rates of full-fledged WWTP applying primary and secondary treatment, which are available from the data introduced by the user. This has the advantage that additional input data in terms of removal rates for primary treatment are not required, thus reducing the data collection burden for the user.

Infrastructure requirements for primary treatment are considered as 30% of an entire WWTP (including all tanks, buildings, anaerobic digester, etc.) in accordance with the inventory processes available in the Ecoinvent 3.2 database. This infrastructure demand is a rough estimate and it is based on the assumption that infrastructure demands are proportional to the overall area requirements of a WWTP. A plant applying only primary treatment requires roughly 70% less area than a complete WWTP, given that elements such as biological reactors, settling tanks, and anaerobic digesters are not needed. These assumptions are incorporated as parameters in the model and the user can change the default parameter values if better data should become available. The electricity requirement for primary treatment was interpolated from the detailed energy consumption reported for US WWTPs by (Stillwell, 2010). All the parameters and data related to primary treatment are provided in the Supplementary Information (SI) in Table S1.

2.2. Tertiary treatment

Small particles of sludge which are not settled in secondary settling tank pass on to the next level of treatment or get discharged with secondary treated wastewater. The fraction of a chemical sorbed to these particles is hence emitted along with these sludge particles, which can be removed by filtration. Sand filtration is considered as the only tertiary treatment in WW LCI 2.0. The inventory for energy requirements and infrastructure requirements of sand filtration was derived from (Muñoz et al., 2010). All parameters and data related to tertiary treatment are provided in the SI in Table S1. The fraction of the chemical adsorbed to the suspended solid phase in the effluent reaching the sand filter can be estimated according to Stuer-Lauridsen et al. (2000):

$$K_d = f_{oc} \times 0.41 \times K_{ow} \quad (3)$$

where:

sK_d is the sludge-water partition coefficients [–]

f_{oc} is the fraction of organic carbon in sludge [–]

K_{ow} , is the octanol-water distribution coefficient of the compound I [–]

In case of dissociating compounds (in this case acids and bases according to the Brønsted–Lowry acid–base definition) D_{ow} should be used instead of K_{ow} and Eq. (3) can be reformulated as:

$$K_d = f_{oc} \times 0.41 \times D_{ow} \quad (4)$$

where:

D_{ow} is the pH corrected octanol-water distribution coefficient of an organic chemical compound accounting for the pH dependency of K_{ow} induced by the presence of ionisable groups in an organic molecule [–]

D_{ow} can be estimated as:

$$D_{ow} = \frac{K_{ow}}{1 + 10^{(pH - pK_a)}} \quad (5)$$

where:

pH is the pH of the effluent of tertiary treatment [–]

pK_a is the acid dissociation constant of chemical compound [–]

Depending on the chemical properties, the fraction of a chemical sorbed to the sludge can be based on either K_d or D_{ow} using the

following equation (Andersen et al., 2004):

$$F_{sludge,tt} = \frac{\rho_{sludge} \times K_d}{1 + \rho_{sludge} \times K_d} \quad (6)$$

where:

ρ_{sludge} is the density (dry solid) of the sludge [g/L].

2.3. Septic tank

Septic tanks are still a common treatment choice in emerging economies (Tilley et al., 2008). Septic tanks are known to remove 30% BOD and achieve 30% removal of SS (Mara, 1996; Crites and Technobanoglous, 1998). Following an approach similar to the one used for WWTPs having only primary treatment, we estimate the chemical-specific degradation and sludge sorption rates by interpolating from the corresponding rates for a WWTP applying primary and secondary treatment, as in Eqs. (7) and (8):

$$F_{deg,st} = r_{deg,st} \times F_{deg,as} \quad (7)$$

$$F_{sludge,st} = r_{sludge,st} \times F_{sludge,as} \quad (8)$$

where, $F_{deg,st}$ is the fraction of chemical removed by biodegradation in septic tanks

$F_{deg,as}$ is the chemical specific fraction of biodegradation in a full-fledged WWTP

$r_{deg,st}$ is the ratio (expressed in percentage) of degradation efficiency in septic tank relative to degradation efficiency in full-fledged WWTP (typically as assumed to be 30%).

$F_{sludge,st}$ is the fraction of solids removed in a septic tank

$F_{sludge,as}$ is the fraction of solids removed in a full-fledged WWTP

$r_{sludge,st}$ is the ration (expressed in percentage) of SS removal efficiency in septic tank to SS removal efficiency in full scale WWTP (typically considered as 30%).

The data related to infrastructure requirements for septic tanks were obtained from Pizzol et al. (2015). All parameters and data related with the septic tank submodel are provided in SI in Table S1. Sludge stored in septic tanks is considered to undergo spontaneous anaerobic digestion before extracted for disposal. In our model this anaerobic digestion is modeled as in WW LCI 1.0 (Muñoz et al., 2016), however biogas generated from this chemical reaction is not collected and escapes to the atmosphere. The septage (the sludge extracted from septic tanks) is assumed to be transferred by a truck and transported (50 km) to a WWTP, where it joins the sludge line. No further chemical transformations are taken into account for the septage once it enters the WWTP, and only energy and infrastructure requirements associated with sludge handling in the WWTP are taken into account.

2.4. Sludge composting

Sludge composting is modeled based on a windrow composting process. The compost plant infrastructure is based on the Ecoinvent data set for a compost plant treating 10,000 ton waste in wet weight per year, with a service life of 25 years (Nemecek and Kägi, 2007). Electricity and diesel use for a sludge composting plant operation was obtained from (Poulsen and Hansen, 2003). The conversion of sludge into compost is based on a mass balance, where only degradable elements in sludge are affected by the composting process. We use the same definition of degradable/inert organic matter as in Muñoz et al. (2016). Dry mass reduction for degradable organic matter is assumed to be 65%. This reduction represents the arithmetic average reduction reported for five fractions commonly found in putrescible organic matter (lignin, cellulose, carbohydrate, fat, protein) in (Sonesson, 1996). Emissions produced by the composting process include CO₂, methane, N₂O, N₂, ammonia and NOx. Methane emissions are calculated as

0.01 kg CH₄/kg degradable organic matter in dry mass (IPCC, 2006), assuming that typical organic matter (food and garden waste) contains 0.435 kg carbon/kg dry mass (IPCC, 2006). Nitrogen in degradable organic matter is emitted as 2% N-N₂O, 2% N₂, 96% N-NH₃ based on (Sonesson, 1996). In addition, we take into account that 15% of the emitted ammonia is converted to NO_x as a secondary pollutant (FAO and IFA, 2001). Thus, the 96% N-NH₃ is in fact modeled as 82% N-NH₃ and 14% N-NO_x. Based on typical values for moisture content in input sludge (Poulsen and Hansen, 2003), wet mass reduction during composting (Poulsen and Hansen, 2003) as well as moisture content in compost (Sonesson, 1996), a fixed water evaporation rate of 70% of the water input is obtained. For mass balance purposes, the consumption of atmospheric oxygen during composting is calculated, based on the stoichiometric requirements to form CO₂ from carbon in degradable organic matter. The amount of final compost obtained in wet weight is calculated with Eq. (9):

$$\text{Compost} = (\text{sludge input} + \text{O}_2) - (\text{water evaporation} + \text{CO}_2 + \text{CH}_4 + \text{N}_2 + \text{N}_2\text{O} + \text{NH}_3 + \text{NO}_x) \quad (9)$$

Based on the initial composition of sludge entering the composting plant and the detailed mass balance, the composition of the resulting compost is obtained, including the fraction (if any) of original chemical released down the drain still remaining in the compost. This composition is used as input for the calculations dealing with application of compost in agriculture, which follows the same principles as sludge application in agriculture in accordance with WW LCI 1.0 (Muñoz et al., 2016).

2.5. Country database

WW LCI 2.0 provides a database on the current status of wastewater treatment levels and sludge disposal practices in 56 countries, based on a wide variety of public sources such as Eurostat, OECD statistics as well as country-specific statistics. Data for 56 countries, on population fractions connected to urban wastewater collection systems - without treatment (%), with primary treatment (%), with secondary treatment (%) and with tertiary treatment (%) are included. Also, population fractions connected to independent wastewater collecting systems with treatment (%) and without treatment is available in the database. For sludge management the percentage of sludge being sent for agriculture, composting, incineration or landfill is also available for all the 56 countries. These statistics are automatically loaded as input data to the model, thus providing a basis for country-specific LCIs. Nevertheless, an option is provided for the user to override these default data in case more accurate data should become available.

2.6. Application to five chemicals

In order to show its applicability in practice, we have assessed five chemicals with WW LCI 2.0. Four chemicals (Ibuprofen, Acetaminophen, DTPMP and Diclofenac) are commonly used in PPCPs and the fifth chemical is Atrazine, which is commonly used as herbicide. The occurrence of these five chemicals in trace amounts is common in sewage (Muñoz et al., 2008; Jiang et al., 2013; Luo et al., 2014). Table 2 shows the list of chemicals with their key features. These molecules have different chemical compositions, biodegradability and toxicities. To further demonstrate the usefulness of the tool, these five chemicals are assessed in four countries viz. Denmark (DK), United States of America (US), China (CN) and India (IN). This choice of countries allows us to model and assess differences in terms of developed vs. developing countries, as well as differences in levels of wastewater treatment and sludge disposal practices (Table 3).

Table 2
Key features of the five chemicals assessed.

Name	CAS number	Key features
Diclofenac	15307-86-5	Organic, poorly degradable, contains N and fossil C ^a
Ibuprofen	15687-27-1	Organic, degradable, contains fossil C ^a
Atrazine	1912-24-9	Organic, poorly degradable, contains N, contains fossil C ^a
Diethylenetriamine penta (methylene phosphonic acid) (DTPMP)	15827-60-8	Organic, soluble, contain N and P, contains fossil C ^a
Acetaminophen	103-90-2	Organic, degradable, contains N, contains fossil C ^a

^a Assumed for the purpose of the study.

Table S2 in the SI shows the physico-chemical properties of these five chemicals. These physico-chemical properties were used to determine fate in the WWTP by using steady-state WWTP simulation models. Various steady-state and dynamic models are available for modelling WWTPs (Gujer et al., 1999; Grau et al., 2007; Ekama, 2009). We have used SimpleTreat (Franco et al., 2013a, b), the model implemented in the European Union framework for the environmental risk assessment of chemicals, to obtain fate factors of these chemicals in a WWTP applying primary treatment plus biological treatment by means of activated sludge. USES-LCA (Van Zelm et al., 2009), which is included in WW LCI 2.0, is used to estimate the fate of the chemicals in the environment (when emitted to either air, water or soil). These fate factors are provided in Table S3 in SI.

Life Cycle Impact Assessment (LCIA) at mid-point level was carried out by importing the inventories generated by WW LCI 2.0 to Simapro 8.0.4. Four relevant impacts categories were assessed using Simapro 8.0.4. The climate change impact category was assessed by means of the global warming potential (GWP) considering a time horizon of 100 years (Forster et al., 2007). Eutrophication (freshwater and marine) impacts were assessed using ReCiPe method (Goedkoop et al., 2008) and USEtox v2.0 (Rosenbaum et al., 2008) was used to assess Freshwater Ecotoxicity (FET) impacts. Terrestrial ecotoxicity was not included in the assessment since USEtox does not currently cover this impact category.

3. Results and discussion

3.1. Inventory results

Key foreground inventory results obtained from WW LCI 2.0 for the five chemicals assessed in four countries are presented in Table S4 in SI. The inventory results reveal that different chemicals involve different resource (energy and chemicals) consumption in each of the four country scenarios.

In DK and US Diclofenac and DTPMP show a net negative electricity consumption, meaning that treatment of these chemicals produces more energy than what additionally is required by the WWTPs and sludge disposal. This is caused by a relatively high sludge production from treatment of effluents containing these two chemicals combined with the fact that the sludge is combustible, achieving credits from energy recovery in relation to sludge incineration. Presence of the remaining three chemicals induces demand for more electricity than can be produced using sludge incineration in the 4 countries. In China and India lower (i.e. lower than in DK and US) wastewater treatment levels prevails (both in terms of secondary as well as tertiary treatment, see Table 3) entailing lower sludge productions from WWTPs in China and India. This leads to a net demand for electricity (i.e. credits from sludge digestion since sludge incineration does not compensate for the electricity requirement for treatment and sludge disposal). Overall, a chemical that is easily biodegradable (e.g. Ibuprofen), has a higher

Table 3
National wastewater treatment levels and sludge handling practice at four countries assessed.

		DK	US	CN	IN
Wastewater treatment scenario	1 Connection to urban wastewater collecting systems - total (%)	89%	81%	46%	21%
	1.1 Connection to urban wastewater collecting systems - without treatment (%)	0%	6%	13%	0%
	1.2 Connection to urban wastewater collecting systems - with treatment (%)	89%	76%	33%	21%
	1.2.1 Connection to urban wastewater treatment - primary treatment (%)	2%	6%	0%	0%
	1.2.2 Connection to urban wastewater treatment - secondary treatment (%)	3%	29%	33%	21%
	1.2.3 Connection to urban wastewater treatment - tertiary treatment (%)	84%	41%	0%	0%
	2 Connection to independent wastewater collecting systems - total (%)	11%	19%	54%	79%
	2.1 Connection to independent wastewater collecting systems - with treatment (%)	11%	19%	0%	39%
Sludge scenario	2.2 Connection to independent wastewater collecting systems - without treatment (%)	0%	0%	54%	39%
	Sludge to composting (%)	6%	12%	4%	0%
	Sludge to landfarming (%)	50%	41%	52%	100%
	Sludge to landfilling (%)	0%	32%	40%	0%
	Sludge to incineration (%)	44%	16%	4%	0%

electricity demand compared to a chemical which is non-biodegradable since it undergoes biological treatment and hence consumes more electricity for e.g. aeration in a WWTP with secondary treatment.

For the treatment of wastewater in cold climate countries such as Denmark there is a heat requirement for maintaining optimal temperatures in the aeration tank. This requirement induces the heat consumption, which can be supplied using co-generation of heat and electricity through sludge incineration. The net balance between the heat required by the treatment process and heat produced using co-generation defines whether there will be positive consumption of the heat or there will be surplus heat generates. Net heat consumption for all chemicals in all countries is found to be negative except for DTPMP in China and India (see Table S4). The reason remains the same as explained for electricity, i.e. the heat produced from sludge digestion and incineration does not compensate for the heat required to operate the WWTP and sludge digestion in CN and IN. This is because the sludge generated by the biodegradable chemicals (e.g. Ibuprofen and Acetaminophen) consists of biomass with a high moisture content (intracellular water) which is not removed by dewatering, whereas non-degradable chemicals such as DTPMP do not yield higher moisture levels of the sludge. Non-degradable organic chemicals do not result in an increased presence of biomass in the sludge but rather in an increased carbon content of the sludge.

Sludge production is relatively high (0.3 to 0.8 kg dry mass/kg chemical) for chemicals that exhibit partition preference towards the solid phase, such as Diclofenac and DTPMP. For Diclofenac this relates to the presence of tertiary treatment in DK and US, while for DTPMP tertiary treatment is not required to cause partitioning to sludge. Chemicals that are at the same time non-biodegradable and do not tend to partition to the solid phase (e.g. Atrazine) yield lower sludge productions (0.02 kg dry mass/kg chemical).

Wastewater treatment also leads to credits originating from avoided mineral fertilizers in those cases where sludge generated in WWTPs is applied to agricultural land. The amount of substituted fertilizer depends on the N and P content of the chemicals undergoing treatment, the chemical's biodegradability and water-sludge partitioning, the country specific level of wastewater treatment and sludge management practices (i.e. use in agriculture). As it can be seen from Table S4, DTPMP obtains relatively high credits for avoided phosphate fertilizer (about $-0.2 \text{ kg P}_2\text{O}_5/\text{kg chemical}$ in Denmark and US and $-0.16 \text{ kg P}_2\text{O}_5/\text{kg chemical}$ in India). In China, only 33% of the wastewater undergoes treatment and only about 50% of the sludge produced in WWTPs is applied to arable land, the fertilizer credit obtained by DTPMP is low

($-0.09 \text{ kg P}_2\text{O}_5/\text{kg chemical}$). For the remaining chemicals, which do not have P in the molecule still lower nutrient credits are achieved due to nitrogen content in sludge.

Direct CO₂ emissions from the discharge of these chemicals can be grouped into four categories viz., emissions from the WWTP (process emissions due to biodegradation), emissions from sludge disposal, degradation of the chemicals in the environment and long-term storage (e.g. landfill). Emissions from the WWTP are higher for Ibuprofen and Acetaminophen in Denmark and US given that in these countries the population fractions connected to WWTPs are high and because these chemicals undergo degradation during biological treatment. Chemicals partitioning to the solid phase end up in the sludge and therefore exhibit significant CO₂ emissions in relation to sludge disposal (e.g. Diclofenac). In China and India, most of the CO₂ emissions are associated with degradation of chemicals in the environment, as there is a relatively low fraction of the populations connected to wastewater treatment in these countries.

One of the more interesting features of WW LCI 2.0 is that the user can obtain a complete impact assessment profile for the chemical released down the drain. The last three rows in Table S4 show, for each of the 5 chemicals, the respective fractions emitted to air, water and soil. More persistent chemicals that tend to partition to sludge such as DTPMP and Diclofenac appear as emissions to soil in the inventory for those countries where sludge is used in agriculture. Persistent chemicals that do not undergo considerable degradation within a WWTP, such as Atrazine, are mainly released to surface water. Surface water is obviously also the main environmental compartment of emission for all chemicals in China and India, where connection to WWTPs by the population is low. Hence, a further impact assessment of such emissions needs to be carried out to determine the health and ecosystem damages due to emission of these chemicals to ambient water.

3.2. Life cycle impact assessment

Fig. 2 shows the impact assessment results for climate change (CC) for all five chemicals in each country. A direct discharge scenario (0% connection to any type of wastewater treatment) is for comparative purposes shown as reference.

DTPMP shows a value of $1.7 \text{ kg CO}_2 \text{ eq./kg}$ released when wastewater is discharged directly without any treatment. Compared to this value, DK and US show CC values of 0.6 and $0.7 \text{ kg CO}_2 \text{ eq./kg}$ released. These lower values can be explained by the fertilizer credit achieved by DTPMP in these countries as well as to the fact that in the direct discharge scenario DTPMP degradation is expected to form some methane

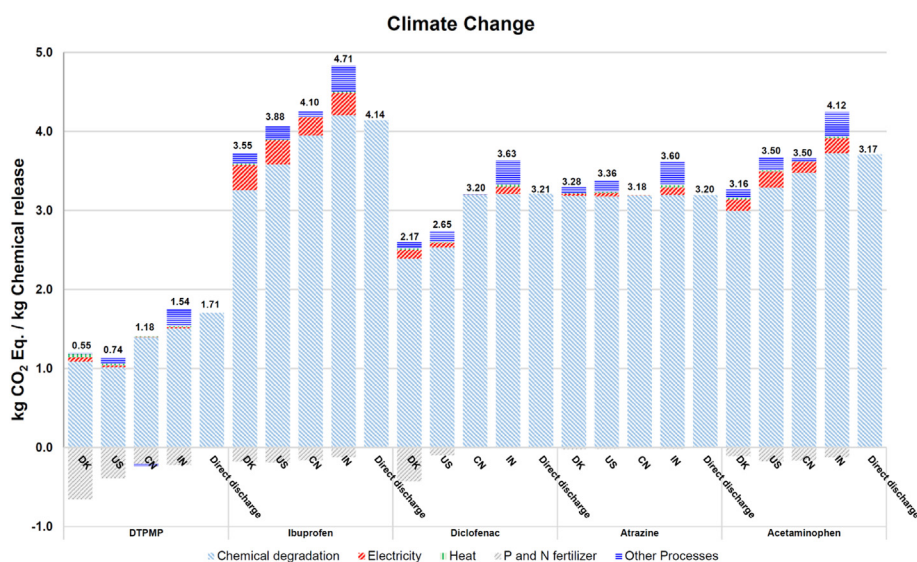


Fig. 2. Impact assessment results for climate change indicator (values on the top of columns represents net climate change value i.e. after deducting credits).

(Muñoz et al., 2016), which is avoided if the chemical passes through WWTPs. In China and India DTPMP cause emissions of 1.2 and 1.5 kg CO₂ eq./kg released. The higher value obtained for India is due to the lower fertilizer credits and higher impact of electricity production used for aeration in WWTPs. The results for the remaining three chemicals (Ibuprofen, Diclofenac and Acetaminophen) can be explained in a similar way. Overall, DK and US exhibit lower CC impact potentials for treatment compared to direct discharge scenarios due to higher levels of secondary and tertiary treatment (and hence higher electricity credits from sludge incineration, see Table S4 in SI). In China and India the lower treatment levels, together with a considerable environmental impacts induced by electricity production due to substantial use of fossil fuels causes the CC impact potentials of the treatment scenarios to equal those for direct discharge. For Atrazine, which is removed neither by secondary nor tertiary treatment, the CC impact potentials in all four countries are almost equal (in DK and CN) or higher (in US and IN) compared to direct discharge scenarios. The reason for this is that there is no removal of Atrazine at any stage of the wastewater treatment, and only little additional impact is induced by infrastructure and energy requirements in the WWTP.

The FET indicator results exhibit a pattern somewhat similar to the CC impact potential as the FET results show similar ranking of the chemicals in each country-specific scenario, except Atrazine (refer to Fig. 3). Overall, Denmark and US obtain lower FET impact potentials compared to China and India for DTPMP, Ibuprofen, Diclofenac and Acetaminophen. Due to its high ecotoxicological impact potential according to USEtox, Atrazine exhibits high FET impact potentials in all the scenarios. For all five chemicals, it is observed that FET results for the direct discharge scenarios are higher than in any of the country specific scenarios including treatment. As expected, wastewater treatment mitigates freshwater ecotoxicity impacts, as long as chemicals are effectively removed by the treatment. The toxicity impact potentials induced by the WWTP's background system (electricity production, infrastructure production, etc.) are lower than the toxicity impacts induced by these chemicals irrespective of wastewater treatment.

The freshwater eutrophication impact potentials (FEP) and the marine eutrophication impact potentials (MEP) are presented in Fig. 4. Chemicals having Nitrogen (N) and/or Phosphorus (P) in their chemical structure exhibit substantially higher FEPs and MEPs. As DTPMP contains 5 atoms of P per molecule and Atrazine contains 5 atoms of N

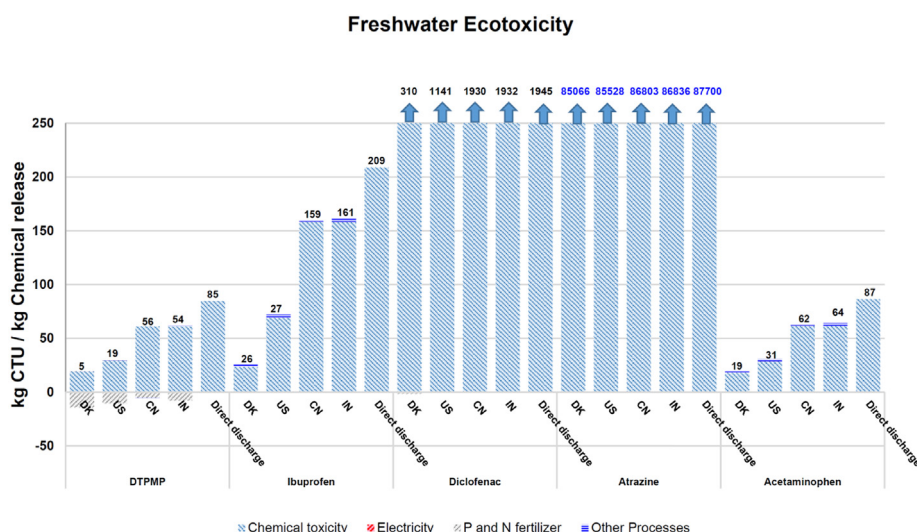


Fig. 3. Impact assessment results for freshwater ecotoxicity (values on the top of columns represents net climate change value i.e. after deducting credits).

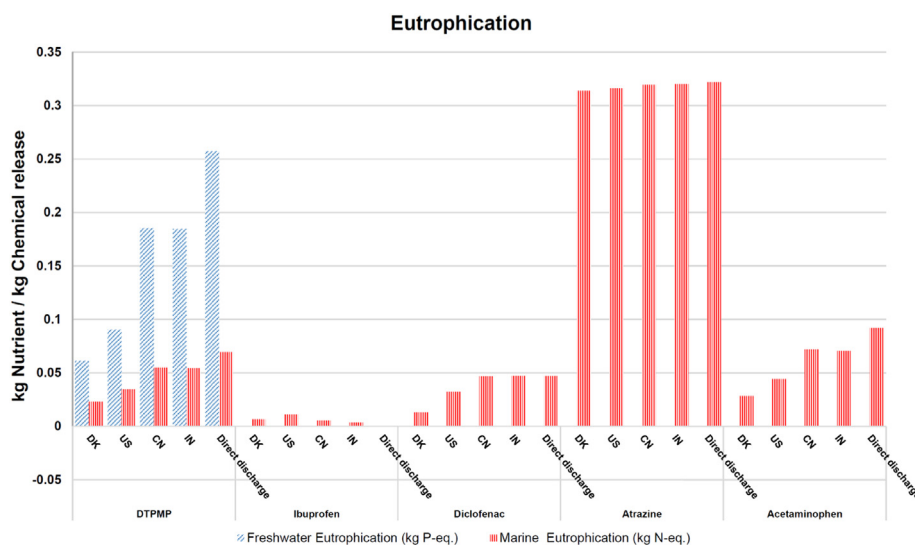


Fig. 4. Impact assessment results for eutrophication.

per molecule, DTPMP and Atrazine yields higher values for FEP and MEP, respectively. Atrazine, Diclofenac and Acetaminophen yields lower FEP compared to DTPMP as these chemicals do not contain P atoms. For all these chemicals, the contribution to FEP originates from the background system (production of energy, auxiliary materials, etc.). It should be highlighted that even though Ibuprofen does not contain N or P, some small contributions for FEPs and MEPs are observed in Fig. 4. These impacts are caused by N in the sludge biomass generated during the biological treatment in the WWTP.

3.3. Uncertainties and limitations of the model

Although the present model goes beyond WW LCI 1.0, it still shares many of its inherent limitations, as discussed by Muñoz et al. (2016). Below we list what in our opinion are the main limitations of WW LCI 2.0:

- The country database on wastewater treatment levels and sludge management practices was derived from various literature sources. These data have a strong influence on the LCIs produced by the model. Although the values reported in the database can be considered as reasonable, the user needs to assess the quality of these data and decide whether or not other more reliable data sources are available.
- Removal efficiencies of organic matter and suspended solids in septic tanks and primary sedimentation tanks are estimated based on literature. These removal efficiencies are then made chemical-specific based on the values for the same chemical in a WWTP using activated sludge. This approach was chosen to minimize the data collection efforts, which for the present version of our model is substantial, but on the other hand constitutes a simple way of modelling the treatment process.
- Tertiary treatment is limited to pressure sand filtration. This is a simplification of reality, since there is currently a wide range of tertiary or advanced treatment methods, such as membrane filtration (from microfiltration to reverse osmosis), disinfection, or advanced oxidation processes (ozonation, photocatalysis). These technologies are not considered in the model.
- Application of wastewater, either treated or untreated, for irrigation of crops or for other uses is not currently included and therefore the model neither accounts for the potential environmental benefits in terms of freshwater resource conservation, nor for the potential impact on terrestrial ecotoxicity derived from chemicals deposited in soil through this emission pathway.

- As opposed to SewageLCI, WW LCI 2.0 does not consider degradation of chemicals in the sewer before they reach the WWTP.
- Management of septage (sludge from septic tanks) is assumed to be treatment in a WWTP. Although this might seem appropriate in the context of a developed country, in developing countries this assumption could be far from reality, given that there are many options currently applied to deal with such waste, from dedicated septage treatment plants to simple discharge of the septage into open drains.
- As in WW LCI 1.0, electricity mixes are country-specific, thus allowing for differentiation of impacts induced by different national electricity mixes. On the other hand, thermal energy production is by default assumed based on natural gas, which may not be realistic in countries like e.g. China, where the use of coal is prevailing.
- In relation to secondary treatment in WWTPs those chemicals that do not contain N in their molecule are assumed to benefit from 'free' N (e.g. N-NH_4^+) available in wastewater, in order to form sludge biomass. However, as a consequence of this N consumption, the treatment of this amount of N by the WWTP (through nitrification and denitrification) is avoided, however this trade-off is not accounted for by the model. This limitation was inherent to WW LCI 1.0 (although not discussed by the authors) and still present in the new version. In this way, the environmental impact of chemicals that do not contain N in their molecule is (slightly) overestimated.

Despite of the above listed uncertainties and limitations related with the model, the WW LCI 2.0 model represents an advancement in the area of wastewater treatment inventories. The model is as far as we know an important applicable missing link between chemical consumption and chemical emission in LCA, especially for chemicals present in products with wide-dispersive use patterns such as e.g. PPCPs.

4. Conclusions

In this paper we have presented WW LCI 2.0, a second-generation model resulting from the integration of two existing models, namely SewageLCI 1.0 and WW LCI 1.0. The purpose of this model is to provide LCIs for specific chemicals under average treatment practices in different countries, and not the assessment of wastewater treatment technologies. The new integrated model generates chemical-specific LCIs of chemicals released to the sewer systems, according to country-specific conditions in terms of wastewater treatment levels as well as typical

sludge disposal practices. The resulting inventories take into account the mass balance of the chemical undergoing treatment and all the required inputs from technosphere (energy, chemicals, infrastructure, etc.). The applicability of this model has been demonstrated by applying it to five chemicals commonly occurring at trace levels in wastewater.

Each of the five chemicals assessed yielded quite different inventories and hence different impact profiles owing to the fact that different chemicals will have different level of wastewater treatment demands. The major findings of our work are:

- i. Similar chemicals yield different inventories in the country specific scenarios, and it has hence been illustrated that different treatment strategies and trade-off options will yield different inventories also for similar chemicals.
- ii. The energy required for wastewater treatment of biodegradable chemicals can be compensated for by the electricity produced from sludge incineration.
- iii. For highly persistent chemicals like Atrazine, indirect toxicity related impacts from treatment systems (including electricity production, production of chemicals, infrastructure, etc.) are lower than direct toxicity related impacts, exerted by these chemicals, when emitted to the environment after treatment.
- iv. Treatment of wastewater is recommended up to the tertiary level in order to reduce environmental impacts such as eco-toxicity and eutrophication, but also climate change.
- v. The model also proves the relevance of sludge disposal practices in terms of the environmental impact trade-offs.

In spite of its limitations, WW LCI 2.0 constitutes a considerable step forward in the modelling of life cycle inventories for chemicals, allowing LCA practitioners to get a more realistic picture of how chemicals contribute to the environmental impacts caused by wastewater discharges.

Acknowledgements

The first author acknowledges H. C. Ørsted Cofund Postdoctoral fellowship received from the People Programme (Marie Curie Actions) of the European Union's Seventh Framework Programme (FP7/2007–2013) under REA grant agreement no. 609405 (COFUNDPostdocDTU). Authors also acknowledge detailed comments received from two anonymous reviewers which helped us to improve the clarity of the manuscript.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2017.10.051>.

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